

SYNTHESIZING A NEW TYPE OF MULLITE LINING

SINTEZA NOVE VRSTE OBLOGE IZ MULITA

Zagorka Aćimović¹, Anja Terzić², Ljubiša Andrić³, Ljubica Pavlović³,
Marko Pavlović¹

¹University of Belgrade, Faculty of Technology and Metallurgy, Belgrade, Serbia

²Institute for Materials Testing, Belgrade, Serbia

³Institute for Technology of Nuclear and Other Mineral Raw Materials, Belgrade, Serbia
anja.terzic@institutims.rs

Prejem rokopisa – received: 2013-03-07; sprejem za objavo – accepted for publication: 2013-03-27

Various possibilities for developing new mullite-based refractory linings that can be applied in a casting process were investigated and are presented in this paper. An optimization of the refractory-lining composition design with the controlled rheological properties was achieved by applying different lining components and altering the lining-production procedure. Mullite was used as a high-temperature filler. A mullite sample was tested with the following methods: X-ray diffraction analysis, differential thermal analysis and scanning-electron microscopy. The particle shape and particle size were analyzed with the program package for an image analysis called OZARIA 2.5. It was proved that an application of this type of lining has a positive effect on the surface quality, structural and mechanical properties of the castings of Fe-C alloys obtained by casting into sand molds, according to the method of expandable patterns (the EPC casting process).

Keywords: refractory lining, mullite, quality of casting, EPC casting process

V članku so predstavljene različne možnosti za razvoj nove mulitne ognjevdružne obloge, uporabne pri postopku ulivanja. Optimiranje sestave ognjevdružne obloge s kontroliranimi reološkimi lastnostmi je bilo doseženo z uporabo različnih sestavin obloge in s spremembo postopka izdelave obloge. Mullit je bil uporabljen kot visokotemperaturno polnilo. Vzorec mulita je bil preiskovan z naslednjimi metodami: z rentgensko difrakcijo, diferenčno termično analizo in vrstično elektronsko mikroskopijo. Oblika in velikost zrn sta bili določeni s programsko opremo za analizo slik OZARIA 2.5. Dokazano je bilo, da uporaba te vrste obloge ugodno vpliva na kvaliteto površine, strukturo in mehanske lastnosti ulitkov iz Fe-C zlitin pri ulivanju v pečene forme po metodi ekspanzirane pene (EPC-postopek ulivanja).

Ključne besede: ognjevdružna obloga, mulit, kvaliteta ulitkov, EPC-postopek ulivanja

1 INTRODUCTION

The main role of a lining is the formation of an efficient, unbreakable and firm refractory barrier which separates the sandy substrate from the liquid metal flow. For such a role, certain lining properties are required: high refractoriness, suitable gas permeability, simple application, good adhesion to a sandy mold and polymer model, simple adjustment of the lining-layer thickness and high drying rate. These requirements can be successfully fulfilled with an optimization of the lining composition and production technology.¹⁻³

The basic characteristic of the EPC casting process is that the patterns and gating of molds made of polymers stay in the cast until the liquid-metal inflow occurs. The pattern-decomposition kinetics is the function of the liquid-metal temperature, with which the pattern comes in contact. The important factors influencing the pattern decomposition and, consequently, the evaporation process, besides temperature and pattern density, are: type of refractory lining, thickness of the lining layers covering the evaporable pattern, type and size of the sand grains and their granulation, permeability of the sandy model, gating of the mold construction, etc.⁴⁻⁷ Manufacturing the castings with the projected application quality by means of the EPC process has not been investigated enough and, thus, there is a need for a systematic

research of the 'triad' including structure/properties/technology, to which special attention was paid in this paper.

The mullite was chosen as the refractory-lining filler due to the following properties: low thermal conductivity ($6 \text{ W m}^{-1} \text{ K}^{-1}$);⁸ low coefficient of the linear thermal expansion ($5.4 \cdot 10^{-6} \text{ }^\circ\text{C}^{-1}$ at $25 \text{ }^\circ\text{C}$);⁹ high thermal-shock resistance (quenching/500) and high maximum-use temperature of $1650 \text{ }^\circ\text{C}$;¹⁰ flexural strength of 180 MPa ; elastic modulus of 151 GPa ; compressive strength of 1310 MPa ; hardness of 1070 kg mm^{-2} ; extreme resistance to liquid-metal absorption;⁶ no gas production when in contact with liquid metal. Different additive types and various quantities were tested in order to enable the best possible absorption between the additives and the refractory filler particles and, thus, the maintenance of the filler in a dispersed state and prevention of the filler build-up or segregation.

2 EXPERIMENTAL WORK

For the synthesis of mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) the mixture of kaolin and alumina was used with an addition of a mineralizer (1 % of NaF). Alumina was added in order to achieve the mullite stoichiometric ratio of 3 : 2. The crushing of the reactive components and the homo-

genization were performed in a planetary ball mill PM 100 with sintered, aluminium-oxide grinding balls. After the homogenization and an addition of the mineralizer, the mixture was wetted with water and, subsequently, pressed in the mold with the 100 N/mm² pressure and, afterwards, dried. The synthesis of mullite was performed by means of isothermal heating in the laboratory high-temperature 'Netzsch' furnace at the temperature of 1450 °C, with the heating rate of 10 °C/min in air atmosphere.

The lining compositions were defined (Table 1) and the lining-component preparation methods were determined.

Table 1: Composition of refractory mullite-based linings

Tabela 1: Sestava ognjevdružne obloge na osnovi mulita

Component	Refractory lining based on alcohol	Refractory lining based on water
ararsid14156519 Refractory filler	Mullite with the grain size of 35–40 µm, 90–94 %	Mullite with the grain size of 40 µm, 93–95 %
Binding agent	Colophonium (C ₂₀ H ₃₀ O ₂), 2.5 %	Bentonite, 2.5%; Bindal H, 0.5%; Na ₃ P ₃ O ₃ , 1–3 %
Additive/suspension	Bentone 25, 0.8–1 %	Carboxymethyl-cellulose, 0.5–1 %
Solvent	Alcohol	Water

Refractory linings were applied to the sandy molds with a brush. During the application of the refractory lining on the polymer model using the technique of immersion into the tank with a lining, the process parameters were: the suspension density of 2 g/cm³, and the suspension temperature of 25 °C. The drying procedure was as follows: for the water-based linings, the duration of drying the first layer was 2 hours and the final layer was dried for 24 h; for the alcohol-based linings burning was used. The thickness of the lining layer on the model after drying was 0.5–1 mm.

For casting, Fe-C alloys were used. The casting temperature was 1350 °C. For the production of sandy molds, the mold mixture based on quartz sand was used, with the mean grain size being 0.17 mm, with an addition of bentonite (3 %) and dextrin (0.5 %). To produce the EPC-casting-process molds, dry quartz sand with the mean grain size of 0.25 mm was used and evaporable models were made of polystyrene with the density of 20 kg/m³.

The mineral-phase composition of mullite was analyzed by means of X-ray powder diffraction (an XRD-Philips PW-1710 diffractometer). DTA was performed with a Shimadzu DTA-50 apparatus. The microstructure of the samples was characterized with the scanning-electron-microscopy method (SEM) using a JEOL JSM-6390 Lv microscope. Distribution of the refractory filler and bonding agent in a lining suspension was conducted with a polarized-light optical microscope of the passing-light JENAPOL type (Carl Zeiss – Jena). The analysis of the

particle size and shape factor was conducted with the PC software OZARIA 2.5.

3 RESULTS AND DISCUSSION

In Figure 1 the results of the X-ray structural analysis of mullite powder are shown. The mean grain size of the refractory filler was between 35–40 µm, the grain-shape mean factor was 0.63, which means that the grains are round and suitable for the production of homogeneous linings.

A DTA curve for the mullite sample is presented in Figure 2. It can be concluded that mullite has a high refractoriness and, thus, it is suitable for casting Fe-C alloys.

In Figure 3, the results of the qualitative mineralogical analysis of the filler based on mullite are shown. The analysis shows that the mullite particles are principally of equal size and morphology, but there are also some differences in the particle size. This is favorable

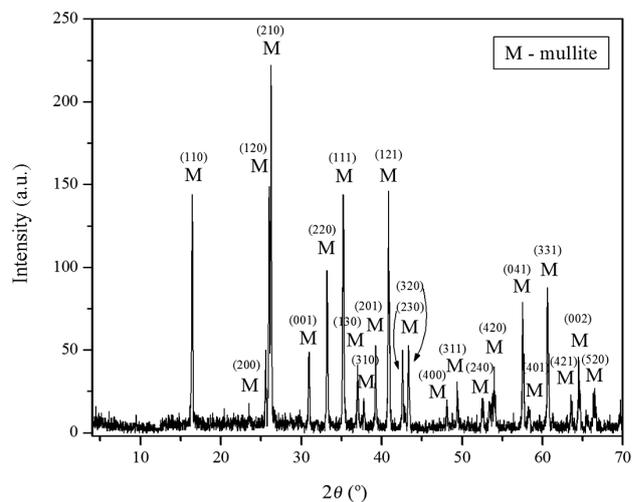


Figure 1: X-ray diffractogram of mullite
Slika 1: Rentgenski difraktogram mulita

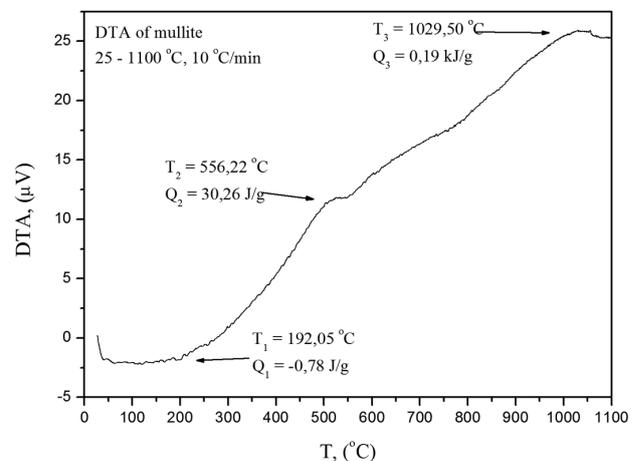


Figure 2: DTA curve of mullite
Slika 2: DTA-krivulja mulita

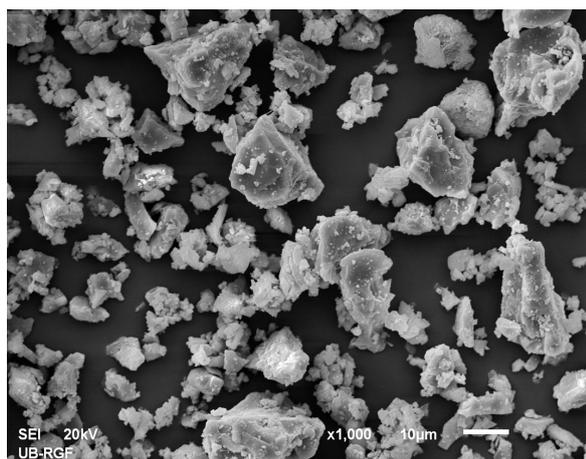


Figure 3: Mullite particles (SEM)
Slika 3: SEM-posnetek zrn mulita

since the particles of diverse granulations contribute to forming an equalized, continuous lining layer on the polymer pattern, due to a better harmony between the particles.

A histogram of the mullite-powder particle size is given in **Figure 4**, while **Figure 5** represents a histogram of the mullite-particle shape factor.

Microstructural analyses of the lining filler and the suspension samples proved that the filler particles have predominantly a uniform size and morphology. The filler particles were mainly oval (**Figure 5**). The filler particles with the sizes of 20–40 µm were predominantly present, making 50 % of the total mullite-particle granulate (**Figure 4**). It was estimated that the oval-shaped particles of various grain sizes contribute to the forming of a uniform and consistent lining layer on the mold and model surfaces due to the stronger interrelations among the particles, which was proved with the results of the lining-property tests. The sediment-stability test performed on all three types of refractory linings showed that solid

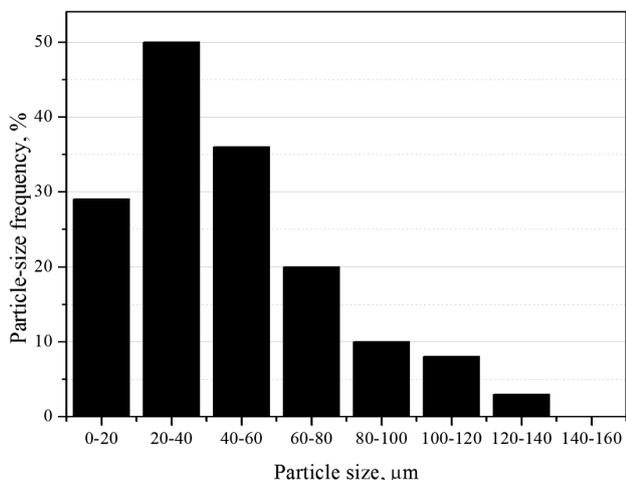


Figure 4: Mullite-particle size frequency
Slika 4: Velikostna razporeditev mulitnih zrn

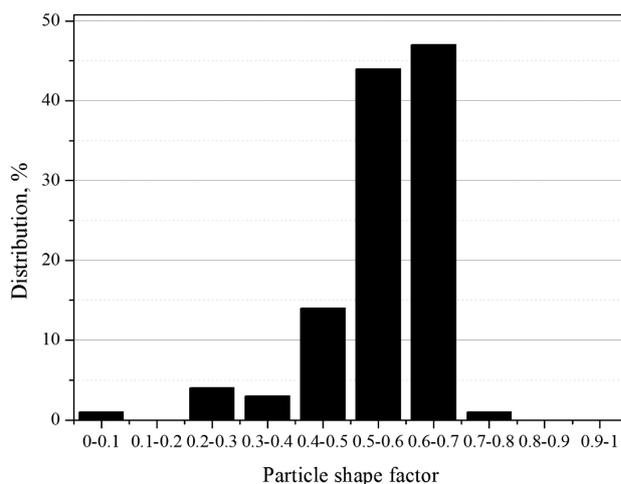


Figure 5: Distribution of the particle-shape factor of mullite
Slika 5: Razporeditev faktorja oblike zrn mulita

matters build up to the amount of 5–8 %, which is in accordance with the lining-quality requirements.

The homogeneity of the refractory-filler distribution also depends on the suspension preparation and application technology. The filler-particle concentration and the adhesion have a significant influence on the suspension rheological properties. In the case of an increase in the concentration of the filler in suspension, it was established that the adhesion forces among mullite particles were also increased and that, under the influence of rheological additives and binding agents, constant and uniform coating layers might be formed on the applied surfaces. The lining adheres easily to the surfaces applied and does not get cracked or wiped out after drying, which was proved after a visual examination of several test samples (**Figure 6**).

An application of diluted linings or the linings whose components are not homogenized enough with careful stirring does not create a good surface adhesion. Furthermore, in this case dried lining layers are not uniform, while the linings applied in thicker layers often crack after drying (**Figure 7**).



Figure 6: Appearance of the mullite-based lining on a test sample without visible flaws

Slika 6: Videz obloge na osnovi mulita na preizkusnem vzorcu brez vidnih napak



Figure 7: Appearance of the mullite-based lining on a test sample with visible flaws

Slika 7: Videz obloge na osnovi mulita na preizkusnem vzorcu z vidnimi napakami

Due to the results of preliminary testing and visual examination of the samples, further tests were carried out using a lining with the density of 2000 kg/m³. The lining components were carefully stirred during the application in order to achieve a homogeneous filler distribution in the suspension.

The experiment showed that alcohol-based linings are suitable for an application in sandy molds and cores. Both alcohol-based and water-based linings applied did not penetrate the test-tube surfaces made of polystyrene.

Applications of the three types of linings, during the processes of casting, in the sandy molds, and the EPC casting procedure enable a production of the castings with advanced quality settings. The prepared linings with a 2000 kg/m³ density were applied in two layers and, after drying, it was noted that constant, thin lining layers were formed on the molds and polymer models. This method provided a good gas permeability of the lining layers, enabling a faster liquid-metal cooling in the mold and a formation of a tiny-grain cast structure, confirmed with the results of testing the structural and mechanical properties of the castings. The application of thin lining layers positively influences the porosity reduction of the castings.

4 CONCLUSION

The investigation showed that the linings based on synthesized mullite met the casting-application requirements creating an acceptable casting-surface finish. This justifies the applied sintering regime at 1450 °C for the mullite synthesization. The investigated linings showed a possibility of an easy application on the sandy molds and polymer models: the linings evenly flew down during the pouring and they were easily applied with a brush without leaving traces, a leakage or a formation of coat drops or lumps. After drying, the lining surfaces were smooth, the layers had an even thickness and the models did not show any bubbling, cracks or peeling. The linings added stiffness to the cluster and allowed the foam-decomposition products to escape. The application of the linings

in thinner layers, approximately 0.5–1 mm, due to improved permeability, showed that the cast quality was higher, having no visible flaws (porosity, bubbles or cracks). Although all the investigated refractory linings gave satisfying properties, water-based linings are more ecologically and economically sustainable than alcohol-based linings.

Further research on improving this type of refractory linings will be done in terms of improved properties of the mullite-based filler using a mechanical activation process enhancing an improvement of the lining rheological properties and the lining suspension stability. Also, further research of the types of refractory linings investigated in this paper will concentrate on the mechanical properties of metal castings in order to confirm the connection between the mechanical performances and the advanced morphological characteristics of the linings.

Acknowledgements

This investigation was supported and funded by the Ministry of Education, Science and Technological Development of the Republic of Serbia and it was conducted within projects 33007, 172057 and 45008.

5 REFERENCES

- ¹ F. G. Martins, C. A. Silva de Oliveira, Study of full-mold casting process for Al–Si hypoeutectic alloys, *Journal of Materials Processing Technology*, 179 (2006), 196–201
- ² M. R. Barone, D. A. Caulk, A foam ablation model for lost foam casting of aluminum, *International Journal of Heat and Mass Transfer*, 48 (2005), 4132–4149
- ³ H. Gökce, D. Agaogullari, M. Lütfi Övecoglu, I. Duman, T. Boyraz, Characterization of microstructural and thermal properties of steatite/cordierite ceramics prepared by using natural raw materials, *Journal of the European Ceramic Society*, 31 (2011), 2741–2747
- ⁴ R. E. Moore, *Refractories, Structure and Properties*, Encyclopedia of Materials: Science and Technology, Press, Oxford 2001, 8079–8099
- ⁵ A. Prstić, Z. Aćimović - Pavlović, S. Grujić, M. Đuričić, Lj. Andrić, Different ceramic linings for application in foundry, *Proceedings of the 43rd October Conference on Mining and Metallurgy, Kladovo*, 2011, 79–82
- ⁶ J. Stjernberg, B. Lindblom, J. Wikström, M. L. Antti, M. Odén, Microstructural characterization of alkali metal mediated high temperature reactions in mullite based refractories, *Ceramics International*, 36 (2010) 2, 733–740
- ⁷ U. Steinhauser, W. Braue, J. Göring, B. Kanka, H. Schneider, A new concept for thermal protection of all-mullite composites in combustion chambers, *Journal of the European Ceramic Society*, 20 (2000) 5, 651–658
- ⁸ G. Brunauer, F. Frey, H. Boysen, H. Schneider, High temperature thermal expansion of mullite: an in situ neutron diffraction study up to 1600 °C, *Journal of the European Ceramic Society*, 21 (2001), 2563–2567
- ⁹ N. M. Rendtorff, L. B. Garrido, E. F. Aglietti, Thermal shock resistance and fatigue of Zircon–Mullite composite materials, *Ceramics International*, 37 (2011) 4, 1427–1434
- ¹⁰ N. Rendtorff, L. Garrido, E. Aglietti, Mullite–zirconia–zircon composites: Properties and thermal shock resistance, *Ceramics International*, 35 (2009), 779–786